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The Strength of Single Crystal Copper under Uniaxial Shock Compression at Mbar Pressures

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In situ x-ray diffraction has been used to measure the shear strain (and thus strength) of single crystal copper shocked to Mbar pressures along the [001] and [111] axes. These direct shear strain measurements indicate shear strengths at these ultra-high strain rates (of order $10^9\mathrm{s}^{-1}$) of a few GPa, which are both broadly in agreement with the extrapolation of lower strain-rate data and with non-equilibrium molecular dynamics simulations.

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Despite many decades of study, the response of materials under shock compression at ultra-high strain rates (10^6 to $10^{10}\,\mathrm{s}^{-1}$) remains poorly understood. In particular, whilst it is known that for many materials the shear stress that can be supported increases with plastic strain rate, $\dot{\epsilon}_\mathrm{p}$, such measurements have largely been limited to relatively modest values of $\dot{\epsilon}_\mathrm{p}$. For example, in the case of copper - the material of study here - Follansbee and Gray used a Kolsy-Hopkinson bar technique to measure shear stress at $\dot{\epsilon}_\mathrm{p}$ between 10^3 and $10^5\,\mathrm{s}^{-1}$,[1] whilst Tong and co-workers extended the range to just above $10^6\,\mathrm{s}^{-1}$ by use of a pressure-shear technique[2] and Meyers and co-workers reached rates of $10^7\,\mathrm{s}^{-1}$ using laser induced shocks.[3] However, above these strain-rates direct measurements of metallic strength have hitherto been inaccessible. The issue has not been one of subjecting material to such high values of $\dot{\epsilon}_\mathrm{p}$ – in their classic paper Swegle and Grady note that within a steady shock $\dot{\epsilon}_\mathrm{p}$ for a large range of materials scales as the fourth power of the peak applied stress,[4] and an extrapolation of their results for copper at low strain rates ($<10^7\,\mathrm{s}^{-1}$) indicates that $\dot{\epsilon}_\mathrm{p}$ of order $10^9\,\mathrm{s}^{-1}$ will be achieved for shock pressures below a Mbar – a pressure region which can readily be accessed. The difficulty in experimentally assessing material strength at high $\dot{\epsilon}_\mathrm{p}$ and high pressures has been the lack of direct experimental techniques for making such measurements (although some data has been obtained in shock-release measurements).[5, 6] Furthermore, VISAR measurements at quite high pressures indicate that the yield stress of aluminium may rise to around several GPa for shocks up to 0.7 Mbar, but the error bars in this regime are extremely large.[7] From the theoretical standpoint, non-equilibrium molecular dynamics (NEMD) calculations of the shear strength of copper at ultra-high strain-rates, in excess of $10^{10}\mathrm{s}^{-1}$, indicate that a shear stress over a GPa can be supported.[8] Importantly, these NEMD simulations are consistent with an extrapolation of the lower strain rate data, and an experimental verification of these results would provide an important demonstration of the validity of the scaling between material strength and strain rate over between 6 and 7 orders of magnitude.

Time resolved x-ray diffraction from shocked materials is a technique that has emerged over recent years as an important tool in shock physics.[9–14]. Importantly, it affords the possibility of providing direct information about material strength, but by measuring shear *strain*, rather than shear stress. Some types of defects may also shift the position of the Bragg peaks, but despite the relatively high defect densities anticipated, this correction is expected to be minor.[15, 16] Thus, invoking the normal assumption that stresses are supported by elastic strains (and assuming zero plastic dilatation), under bulk uniaxial compression simultaneous measurements of the lattice parameters in directions perpendicular and parallel to the shock propagation direction provide a direct measure of shear strain, volumetric compression, and the elastic and plastic components of strain along both these directions.

To date, all measurements using in situ diffraction to study shock compressed matter have been limited to shock pressures of order 320 kbar or less.[17] An extension of this pressure range to the Mbar regime would not only open up the range of strain-rates that can be studied in shocked materials, as noted above, but also would demonstrate the viability of the x-ray technique for obtaining information about the crystal lattice under transient shock conditions at pressures rivaling those that can be achieved in diamond anvil cells.

In this letter we report the first direct measurements of shear strain in single crystal copper at shock pressures in excess of a Mbar. We demonstrate a shear strength at these ultra-high strain rates (of order $10^9\,\mathrm{s}^{-1}$) which is both broadly in agreement with the extrapolation of the lower strain-rate data and with non-equilibrium molecular dynamics simulations, and note that for single crystal copper the observation of shear strain is more readily achieved for shocks propagating along [001], rather than [111], owing to the much smaller shear modulus in this direction.

The experiments were performed in Target Area East of the VULCAN high-power laser system[18] at the Rutherford Appleton Laboratory in the UK. Samples of $10\,\mu$m thick, single crystal copper, 5 mm in diameter, were coated with with $19.5\,\mu$m of CH and then $0.3\,\mu$m of aluminum. These samples were shock loaded by direct laser radiation of the Al-coated side of the target, using 6 ns pulses of $1.053\,\mu$m laser radiation in a laser spot of diameter approximately 2 mm. These pulses had a rise time of 150 ps to a maximum value, after which there was a fall off to around half that value, followed by a linear fall within 200 ps. The energy in the laser beams could be varied up to a total 1250 J, providing an average irradiance of up to almost 10^{13} Wcm^{-2}.

The stress along the shock propagation direction was measured by recording the free-surface velocity of the rear surface (i.e. opposite to the irradiated surface) of the single crystal by use of a twin bed line VISAR system. The VISAR signals were recorded on streak cameras with a sweep speed of 1 ns/mm, which gave a time window of order 20-25 ns. On one bed a 28.8 mm etalon was used giving a velocity per fringe of $1.729\,\mathrm{kms}^{-1}$ with temporal resolution of 150 ps and on the second bed a 50 mm etalon was used resulting in a velocity per fringe of $0.996\,\mathrm{kms}^{-1}$ with a temporal resolution of 260 ps.

Simultaneously with the VISAR measurements, the state of strain within the shocked crystals was monitored by in situ divergent beam x-ray diffraction.[19] Quasi-monochromatic Iron He-α x-rays were created by illuminating an iron foil of order 1 mm from the crystal surface with a 1 ns, 150 J pulse of 527 nm wavelength laser radiation, focussed to a $20\,\mu$m spot. X-rays diverging from this point source irradiated the crystal at a range of angles, being diffracted when they match the appropriate Bragg condition, and recorded on large area image plate detectors placed several cm from the x-ray source and crystal. The crystals were sufficiently thin to allow the diffraction patterns to be recorded simultaneously in both reflected and transmitted geometries. The x-ray source with respect to a reference point on the crystal was positioned to within $\pm 20\,\mu$m. Fitting the positions of the detectors relative to the crystal to multiple lines from the unshocked crystal means that the dominating source of error is the original crystal quality and the finite bandwidth of the x-ray source. Furthermore, this alignment procedure allowed us to determine the position on the surface of the crystal from which x-rays were being diffracted, with respect to the center of the shock-drive laser beams, to an accuracy of $\pm 50\,\mu$m. The VISAR traces showed that the shock breakout occurs within a 150 ps window over a 1.2 mm region of the crystal, and the position of the x-ray source was set to ensure that the diffracted x-rays used for determining the degree of longitudinal and transverse strain within the shock were scattered from this region of the crystal.

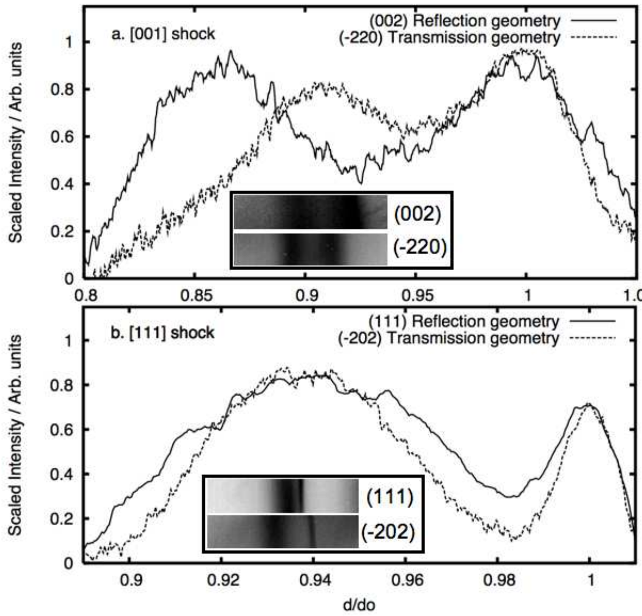

FIG. 1: The intensity profiles for the shocked and unshocked peaks measured in reflection geometry and in transmission geometry and the strain deduced for (a.) [001] shock, where (002) was the reflected peak and (-220) was the transmitted peak, and (b.) [111] shock where (111) was the reflected peak and (-202) was the transmitted peak. Insets show raw data from which the lineouts were taken.

For shocks along the [001] direction the compression in the shock direction was measured from the (002) Bragg peak, whilst on the same shot the strain perpendicular to the shock direction was measured from the (-220) peak. The image plate data is shown in Fig. 1, along with a plot of diffracted intensity against strain for each of the relevant directions. We observe a compression along the shock direction of 13±1%, and 9±1% perpendicular to the shock direction – that is to say a value of V/V_0 of 72%. For shocks along the [111] direction the compression in the shock direction is measured from the (111) peak whilst on the same shot the strain perpendicular to

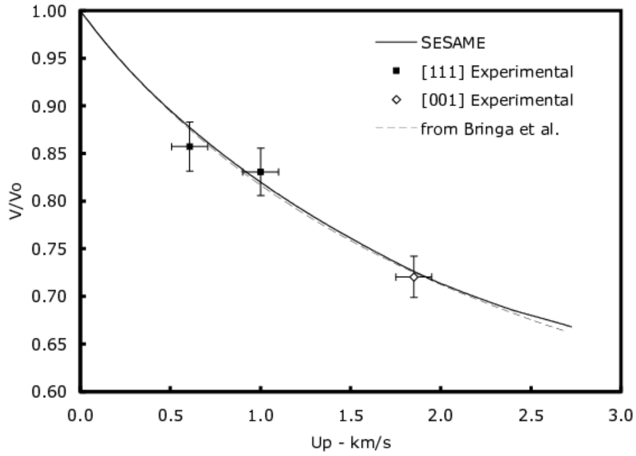

FIG. 2: The comparison of shocks along [111] (squares) and [001] (diamonds) with SESAME equation of state predictions and molecular dynamics simulations[23]. The experimental results from VISAR gives the piston velocity and the diffraction data gave the compressions.

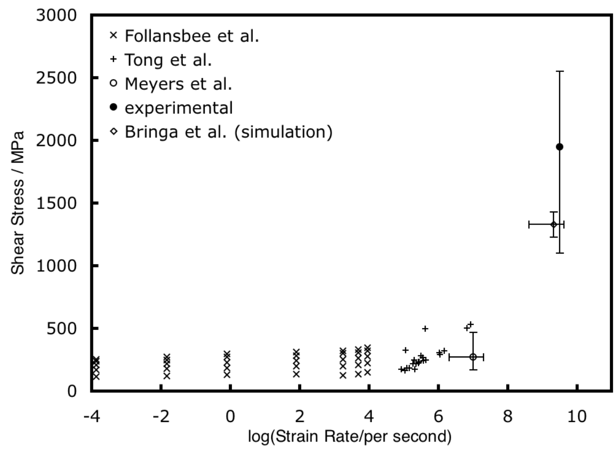

FIG. 3: Shear stress of copper at various strain rates. Data taken from Experiments[2, 3, 21] and current estimates, and from Molecular Dynamics simulations.[8] In the cases where the shear stress was calculated from the shear strain, this was done by simulating an ideal crystal with the appropriate longitudinal and transverse strains

the shock direction was measured from the (-202) peak. In this case it is found that there is no measurable difference between the lattice strains parallel to the shock direction and strains perpendicular to the shock direction up to pressures of 0.5 Mbar, a point to which we shall return later. A summary of the experimental data can be found in table I.

The simultaneously measured VISAR signals allow the compression data deduced from x-ray diffraction to be compared with EOS predictions. Taking the particle velocity just before release to be half the free-surface velocity measured with VISAR, we show in Fig. 2 the experimental diffraction and VISAR data alongside predictions from both the SESAME EOS[20], and the compression-U_p curve deduced by Bringa and co-workers using the LAMMPS molecular dynamics package to simulate shock waves in copper,[22, 23] with the material response being modelled using Mishin's embedded atom model (EAM) potential.[24] Excellent agreement is found.

Defining shear strain as

$$\gamma = \tan\left[2\tan^{-1}\left(\frac{1-\epsilon_1}{1-\epsilon_2}\right) - \frac{\pi}{2}\right] \quad , \tag{1}$$

where ϵ_2 is the compressive strain in the direction in which the shock is travelling and ϵ_1 is the transverse compressive strain. This gives a shear strain of 0.045 at $V/V_0 = 72\%$ for the shock along [001]. It is known that under compression the shear modulus will differ considerably from its zero-pressure value. Indeed, from LAMMPS simulations using the Mishin EAM potential, we infer a shear stress of 1.9 GPa for the [001] shock from which we deduce a shear modulus under this compression of 42 GPa, a factor of 2 higher than that at zero pressure value. In these calculations shear stress is given by

$$\tau = \frac{1}{2}\left[\sigma_{zz} - \frac{1}{2}(\sigma_{xx}+\sigma_{yy})\right] \quad , \tag{2}$$

where σ_{ii} is the stress along the axis i and z is the axis of compression.

Shock Direct.	Long. Strain/%	Trans. Strain/%	Total Comp/%	U_p kms^{-1}	Pressure /Mbar
[001]	13±1	9±1	28±2	1.85±0.05	1.2±0.2
[111]	6±1	6±1	17±3	1.0±0.05	0.5±0.2
[111]	5±1	5±1	14±3	0.6±0.05	0.3±0.1

TABLE I: Summary of atomic strains and particle velocities measured and shock pressures inferred from the SESAME equation of state.[20]

The rate at which the lattice is strained can be deduced from the diffraction data, noting that the diffracted intensity between the peaks corresponding to the uncompressed material, and shocked material can be associated with the finite rise-time of the strain front within the crystal. By iteratively solving the dynamical diffraction equations for a monotonic strain profile, and comparing the diffracted intensity with that found experimentally,[25, 26] we find a shock width of order ***μm, corresponding to a rise time of order ***psec.. That is to say the overall strain rate is ****s^{-1}. The experimentally determined shear stress for this strain rate is plotted alongside the lower strain rate data in Fig. 3. In the same figure we show the shear stress deduced from LAMMPS simulations performed by Bringa and co-workers, where we have again deduced stresses from the published strain data using compression-dependent shear moduli, and deduce the rate from the rate of decay of shear stress. It can be seen that the experimental data and MD simulations at these ultra-high strain rates indeed show high shear stresses, which agree within of order a factor of two.

As noted above, and can be seen from Fig. 1, within the margins of experimental error there is no evidence for a sustained shear strain for those crystals shocked along [111] directions. However, it is important to note that this does not imply that the shear stress is small – the shear moduli along [001] and [111] differ by a factor of 3 even at low compressions, and thus high shear stresses along [111] correspond to quite small shear strains. This large difference in behavior is related to the fact that uniaxial compression of an fcc crystal along [001] takes the crystal along the Bain path which has the effect of keeping the shear stress relatively low. Compressing along the [111] direction has no such moderating influence. To investigate this effect further we used LAMMPS to investigate shock compression along [111] in Cu. A $616\times1067\times4715$Å(259 million atom) single crystal of copper was thermalised to 300K. To generate the shock all atoms within two conventional cells of $z = 0$ were fixed and then driven as a unit into the crystal in the positive z direction at a piston velocity of $1\,\mathrm{kms}^{-1}$. An x-ray diffraction pattern was simulated by taking the fourier transform of the coordinates of the relaxed region behind the shock[27, 28]. The elastic strains deduced from the simulated diffraction indicated a shear strain of 0.01, even though LAMMPS predicted a shear stress of close to 3 GPa – consistent with a larger shear modulus along this direction. That is to say that although the diffraction technique we have outlined here provides good information on supported shear stresses within the material for the [001] direction, its applicability will be limited in cases of large shear moduli.

In summary we have used in situ x-ray diffraction to measure shear strains in single crystals of copper shocked to pressures in excess of a Mbar – a pressure that starts to rival those obtainable in DAC experiments. Simultaneous VISAR measurements allow us to show that the compressions deduced from the diffraction data are in agreement with MD and SESAME tables. We find that a shear stress of order 2 GPa is supported for shocks along the [001] direction, at a strain rate of order $***\mathrm{s}^{-1}$, a figure that is in agreement within a factor of two of relevant MD simulations, and significantly higher than lower strain-rate data. Although MD simulations indicate similarly large shear stresses can be supported along [111], the large shear modulus results in it being more difficult to empirically deduce the shear strain, and hence stress along this direction.

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